

Corrections to Wigner type phase space methods

Wolfgang Gaim¹ and Caroline Lasser²

¹ Fachbereich Mathematik, Eberhard Karls Universität Tübingen, 72076 Tübingen, Germany

² Zentrum Mathematik, Technische Universität München, 80290 München, Germany

E-mail: wolfgang.gaim@uni-tuebingen.de, classer@ma.tum.de

Abstract. Over decades, the time evolution of Wigner functions along classical Hamiltonian flows has been used for approximating key signatures of molecular quantum systems. Such approximations are for example the Wigner phase space method, the linearized semiclassical initial value representation, or the statistical quasiclassical method. The mathematical backbone of these approximations is Egorov's theorem. In this paper, we reformulate the well-known second order correction to Egorov's theorem as a system of ordinary differential equations and derive an algorithm with improved asymptotic accuracy for the computation of expectation values. For models with easily evaluated higher order derivatives of the classical Hamiltonian, the new algorithm's corrections are computationally less expensive than the leading order Wigner method. Numerical test calculations for a two-dimensional torsional system confirm the theoretical accuracy and efficiency of the new method.

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1. Introduction

Molecular quantum systems are described by an unbounded self-adjoint operator, the Schrödinger operator

$$H = -\frac{\varepsilon^2}{2}\Delta + V,$$

acting on the Hilbert space of complex-valued square-integrable functions $L^2(\mathbb{R}^d)$. Here, $\varepsilon > 0$ is a small positive parameter related to the inverse square root of the average nuclear mass, and $V : \mathbb{R}^d \rightarrow \mathbb{R}$ is the nuclear potential function resulting from the Born–Oppenheimer approximation, see [20]. The relevant time scales of nuclear quantum motion are of the order $1/\varepsilon$. The time evolution of an initial wave function $\psi_0 \in L^2(\mathbb{R}^d)$ is governed by the time-dependent linear Schrödinger equation

$$i\varepsilon\partial_t\psi_t = H\psi_t \tag{1}$$

with the appropriate ε -scaling of the time-derivative or – equivalently – by the action of the unitary evolution operator $e^{-iHt/\varepsilon}$, since

$$\psi_t = e^{-iHt/\varepsilon}\psi_0 \quad (t \in \mathbb{R}).$$

Even though the Schrödinger equation (1) is a *linear* partial differential equation, the numerical simulation of physical quantities derived from the wave function ψ_t is notoriously difficult for two reasons: The dimension $d \gg 1$ of the nuclear configuration space is large. If a molecule consists of n nuclei, then $d = 3n$. Just for a single water molecule, for example, we have $d = 3 \cdot 3 = 9$. Moreover, nuclear quantum motion is highly oscillatory. Solutions typically oscillate with frequencies of the order $1/\varepsilon$ in time and space, while ε ranges between 0.001 and 0.1, depending on the molecular system under consideration. For the hydrogen molecule H_2 , for example, one has $\varepsilon \approx 0.0233$, while $\varepsilon \approx 0.0035$ for iodine monobromide IBr.

As an answer to these challenges, chemical physicists have developed approximations involving the *nonlinear* time evolution of classical mechanics. One uses the Hamiltonian function

$$h : \mathbb{R}^d \times \mathbb{R}^d \rightarrow \mathbb{R}, \quad h(q, p) = \frac{1}{2}|p|^2 + V(q), \tag{2}$$

the associated Hamiltonian system

$$\dot{q}_t = \partial_p h(q_t, p_t), \quad \dot{p}_t = -\partial_q h(q_t, p_t), \tag{3}$$

and the corresponding Hamiltonian flow

$$\Phi^t : \mathbb{R}^{2d} \rightarrow \mathbb{R}^{2d}$$

as the classical counterparts to the Schrödinger operator H , the time-dependent Schrödinger equation (1), and the unitary evolution operator $e^{-iHt/\varepsilon}$. This quantum-classical correspondence is most elegantly elaborated by using Wigner functions and Weyl quantization.

The Wigner function $W_\psi : \mathbb{R}^{2d} \rightarrow \mathbb{R}$ of a square integrable function $\psi \in L^2(\mathbb{R}^d)$ is a real-valued, square integrable, continuous function on phase space \mathbb{R}^{2d} obtained by an

inverse Fourier transform of the autocorrelation function of ψ , see the short summary in Appendix A. The Wigner function can be thought of as a probability density on phase space \mathbb{R}^{2d} , though in general it might attain negative values. It has been introduced by E. Wigner in [25] when developing the thermodynamics of quantum mechanical systems. Crucial properties of the Wigner function are the orthogonality relation

$$|\langle \phi, \psi \rangle|^2 = (2\pi\varepsilon)^d \int_{\mathbb{R}^{2d}} W_\phi(z) W_\psi(z) dz \quad (\phi, \psi \in L^2(\mathbb{R}^d))$$

and the asymptotically classical time evolution

$$W_{\psi_t} = W_{\psi_0} \circ \Phi^{-t} + O(\varepsilon^2), \quad \varepsilon \rightarrow 0, \quad (4)$$

for the solution ψ_t of the Schrödinger equation (1). The Wigner phase space method of E. Heller [8, 2] and the statistical quasiclassical method of H. Lee and M. Scully [13], for example, use these properties for computing the transition probabilities from a given state ϕ to ψ_t according to

$$|\langle \phi, \psi_t \rangle|^2 \approx (2\pi\varepsilon)^d \int_{\mathbb{R}^{2d}} W_\phi(\Phi^t(z)) W_{\psi_0}(z) dz.$$

Viewing the Wigner function W_ψ of a square integrable function ψ as a tempered distribution, that is, as a continuous linear mapping from the Schwartz functions $a : \mathbb{R}^{2d} \rightarrow \mathbb{R}$ to the real numbers, one arrives at the identity

$$\int_{\mathbb{R}^{2d}} a(z) W_\psi(z) dz = \langle \psi, \text{op}(a) \psi \rangle, \quad (5)$$

where $\text{op}(a)$ denotes the bounded linear operator on $L^2(\mathbb{R}^d)$ obtained by the Weyl quantization of the function a . The Weyl operator $\text{op}(a)$ is a pseudo-differential operator, a generalized partial differential operator, which treats differentiation and multiplication by functions on the same footing, see the appendix. With an appropriate handling of operator domains, Weyl quantization also applies for unbounded linear operators, and the Schrödinger operator $H = \text{op}(h)$, for example, is the Weyl quantized classical Hamiltonian function h defined in (2). Often, the Weyl operator $\text{op}(a)$ is also called the quantum observable associated with the classical observable a . Besides the convenient relation (5), Weyl quantization enjoys the beautiful property that the trace of the product of Weyl operators can be expressed as the integral

$$\text{tr}(\text{op}(a)\text{op}(b)) = (2\pi\varepsilon)^{-d} \int_{\mathbb{R}^{2d}} a(z)b(z) dz,$$

provided that the classical observables a and b satisfy appropriate regularity and growth conditions, see e.g. [4, Proposition 9.2] or [7, Proposition 284].

From the Weyl point of view, the asymptotic time evolution of the Wigner function (4) can equivalently be formulated as

$$e^{iHt/\varepsilon} \text{op}(a) e^{-iHt/\varepsilon} = \text{op}(a \circ \Phi^t) + O(\varepsilon^2), \quad \varepsilon \rightarrow 0. \quad (6)$$

This quantum-classical approximation of time evolved quantum observables is known as Egorov's theorem and has first been formulated in [5] within the Hörmander theory of pseudo-differential operators, see also [19, 1, 26] for refined error estimates in the context

of semiclassical microlocal analysis. The computational power of classically propagating quantum observables has been recognized by W. Miller and H. Wang [17, 24], who approximate the time-dependent correlation function of $\text{op}(a)$ and $\text{op}(b)$ according to

$$\text{tr}(e^{iHt/\varepsilon} \text{op}(a) e^{-iHt/\varepsilon} \text{op}(b)) \approx (2\pi\varepsilon)^{-d} \int_{\mathbb{R}^{2d}} a(\Phi^t(z)) b(z) \, dz.$$

In the chemical physics literature this approximation is referred to as the linearized semiclassical initial value representation (LSC-IVR), and it is derived from Fourier integral operator representations of the unitary evolution operator $e^{-iHt/\varepsilon}$, see also [22] for a recent review. It seems that the link of LSC-IVR to Egorov's theorem has not been noticed so far.

In a study of the one-dimensional Morse oscillator, H. Lee and M. Scully [14] have proposed to improve the second order approximation (4) for the Wigner function of ψ_t by $W_{\psi_t} \approx W$ with

$$\partial_t W = -p \partial_q W + V'_{\text{eff}} \partial_p W,$$

where the derivative of the effective potential V_{eff} is given by

$$V'_{\text{eff}} = V' - \frac{\varepsilon^2}{24} \gamma V''''.$$

For the correcting factor $\gamma = \gamma(q, p, t)$ they mention the three options

$$\gamma_1 = \partial_p^3(W_{\psi_0} \circ \Phi^{-t}) / \partial_p W, \quad \gamma_2 = \partial_p^3(W_{\psi_0} \circ \Phi^{-t}) / \partial_p(W_{\psi_0} \circ \Phi^{-t}), \quad \gamma_3 = \partial_p^3 W / \partial_p W,$$

and they use the term Wigner trajectories for the solutions of the ordinary differential equation $\dot{q}_t = p_t$, $\dot{p}_t = -V'_{\text{eff}}(q_t, p_t, t)$, see also [11, 12]. Later, A. Donoso and C. Martens [3] have defined their entangled classical trajectories by a fourth variant $\gamma_4 = \partial_p^2 W / W$. Trajectories generated by γ_3 and γ_4 and generalisations thereof have also been incorporated for the numerical simulation of time-dependent correlation functions of one-dimensional systems by J. Liu and W. Willer [15].

Our aim here is also a higher order approximation of the dynamics. However, we will work with the rigorous mathematical framework of Egorov's theorem (6) and construct phase space trajectories without any entanglement for arbitrary dimensions $d \geq 1$. The key element for proving Egorov's theorem is an asymptotic expansion of the commutator

$$\frac{i}{\varepsilon} [\text{op}(h), \text{op}(a)] \sim \sum_{k \in 2\mathbb{N}} \left(\frac{\varepsilon}{2i} \right)^k \text{op}(\{h, a\}_{k+1}). \quad (7)$$

where $\{h, a\}_{k+1}$ denotes a generalization of the usual Poisson bracket involving derivatives of the functions h and a up to order $k+1$, see Section 2. In the context of response theory, M. Kryvohuz and J. Cao [9] use this expansion up to the fourth term for a systematic improvement of linear response computations over long times. The commutator expansion (7) also reveals, that for Hamiltonians h , which are polynomials of degree less or equal than two, the remainder of Egorov's theorem vanishes. This exact Egorov result is utilized by H. Waalkens, R. Schubert and S. Wiggins for their quantum normal form algorithm in dynamical transition state theory [23].

For general Hamiltonian functions h , the expansion (7) implies a higher order version of Egorov's theorem,

$$e^{iHt/\varepsilon} \text{op}(a) e^{-iHt/\varepsilon} \sim \sum_{k \in 2\mathbb{N}} \varepsilon^k \text{op}(a_k(t)) \quad (8)$$

with leading order term $a_0(t) = a \circ \Phi^t$ and corrections

$$a_k(t) = \sum_{l \in \{0, 2, \dots, k-2\}} \left(\frac{i}{2}\right)^{k-l} \int_0^t \{h, a_l(\tau)\}_{k+1-l} \circ \Phi^{t-\tau} d\tau.$$

It is remarkable that the corrections are only built from derivatives of the observable a , the Hamiltonian function h and the flow Φ^t , which has also been emphasized by M. Pulvirenti in [18], when deriving higher order estimates for the Wigner function of the wave function ψ_t .

Since the higher order Egorov expansion (8) is built of even powers of the parameter ε , the first correction

$$a_2(t) = -\frac{1}{4} \int_0^t \{h, a \circ \Phi^\tau\}_3 \circ \Phi^{t-\tau} d\tau \quad (9)$$

provides the fourth order estimate

$$e^{iHt/\varepsilon} \text{op}(a) e^{-iHt/\varepsilon} = \text{op}(a \circ \Phi^t + \varepsilon^2 a_2(t)) + O(\varepsilon^4).$$

The aim of this paper is the exemplary analysis of this correction and its discretization for the numerical computation of expectation values. In a first step, we reformulate it as

$$a_2(t) = \Lambda_1^t(Da \circ \Phi^t) + \Lambda_2^t(D^2a \circ \Phi^t) + \Lambda_3^t(D^3a \circ \Phi^t),$$

where

$$\Lambda_k^t : \mathbb{R}^{2d \times \dots \times 2d} \rightarrow \mathbb{R} \quad (k = 1, 2, 3),$$

is an explicitly defined linear mapping from the space of k -tensors to the real numbers, which depends on derivatives of the Hamiltonian function h and the flow Φ^t , but not on the observable a . In spirit, this formulation of $a_2(t)$ is close to the Wigner trajectories generated by the first correction factor γ_1 . In the next step, we derive a first order ordinary differential system for the components of Λ_1^t , Λ_2^t , and Λ_3^t . The vectorization of this equation results in

$$\frac{d}{dt} \Lambda^t = N^t \Lambda^t + C^t, \quad (10)$$

where the building blocks of the matrix N^t and the vector C^t are components of the tensors D^2h , D^3h , and D^4h evaluated along the flow Φ^t . In the final step we derive a fourth order splitting scheme for the discretization of the ordinary differential equation (10), which is then applied for numerical test calculations.

This paper is organized as follows. Section 2 develops Egorov's theorem to the next order with respect to the parameter ε and formulates this correction as a first order ordinary differential equation. Section 3 discusses a discretization of this

corrected approximation for the computation of expectation values. Section 4 provides numerical experiments for a two-dimensional torsional quantum system, which confirm the theoretical considerations. The appendix summarizes basic properties of Wigner functions and Weyl operators.

2. Higher order Corrections

Let $h : \mathbb{R}^{2d} \rightarrow \mathbb{R}$ be a smooth function of subquadratic growth. That is, for all $\gamma \in \mathbb{N}^{2d}$ with $|\gamma| \geq 2$ there exists $C_\gamma > 0$ with

$$\|D^\gamma h\| \leq C_\gamma.$$

Let $a : \mathbb{R}^{2d} \rightarrow \mathbb{R}$ be a Schwartz function. Then, the following formal considerations can be turned into a proof according to [19, Théorème IV.10] or [1, Theorem 1.2].

We look for an approximate classical observable $a_{\text{appr}}(t) : \mathbb{R}^{2d} \rightarrow \mathbb{R}$ such that

$$e^{iHt/\varepsilon} \text{op}(a) e^{-iHt/\varepsilon} \approx \text{op}(a_{\text{appr}}(t)).$$

We require $a_{\text{appr}}(0) = a$ at time $t = 0$, rewrite the difference according to

$$\begin{aligned} e^{iHt/\varepsilon} \text{op}(a) e^{-iHt/\varepsilon} - \text{op}(a_{\text{appr}}(t)) &= \int_0^t \frac{d}{d\tau} (e^{iH\tau/\varepsilon} \text{op}(a_{\text{appr}}(t - \tau)) e^{-iH\tau/\varepsilon}) d\tau \\ &= \int_0^t e^{iH\tau/\varepsilon} \left(\frac{i}{\varepsilon} [\text{op}(h), \text{op}(a_{\text{appr}}(t - \tau))] - \frac{d}{dt} \text{op}(a_{\text{appr}}(t - \tau)) \right) e^{-iH\tau/\varepsilon} d\tau, \end{aligned}$$

and observe that the commutator

$$[\text{op}(h), \text{op}(a_{\text{appr}}(t))] = \text{op}(h) \text{op}(a_{\text{appr}}(t)) - \text{op}(a_{\text{appr}}(t)) \text{op}(h)$$

plays a crucial rule. Multiplying this commutator with $-i\varepsilon$, we obtain an asymptotic expansion in even powers of ε ,

$$\frac{i}{\varepsilon} [\text{op}(h), \text{op}(a_{\text{appr}}(t))] \sim \sum_{k \in 2\mathbb{N}} \left(\frac{\varepsilon}{2i} \right)^k \text{op}(\{h, a_{\text{appr}}(t)\}_{k+1}), \quad (11)$$

where

$$\{f, g\}_k = \sum_{|\alpha+\beta|=k} \frac{(-1)^{|\beta|}}{\alpha! \beta!} \partial_q^\alpha \partial_p^\beta g \partial_q^\beta \partial_p^\alpha f.$$

denotes the k th generalized Poisson bracket of two smooth functions $f, g : \mathbb{R}^{2d} \rightarrow \mathbb{R}$, see Appendix A. Since the first generalized Poisson bracket coincides with the usual Poisson bracket, $\{f, g\}_1 = \partial_p f \partial_q g - \partial_q f \partial_p g$, we have

$$\frac{i}{\varepsilon} [\text{op}(h), \text{op}(a_{\text{appr}}(t))] = \text{op}(\{h, \text{op}(a_{\text{appr}}(t))\}) + O(\varepsilon^2).$$

Let $\Phi^t : \mathbb{R}^{2d} \rightarrow \mathbb{R}^{2d}$ be the flow associated with the classical Hamilton function h , and set $a_{\text{appr}}(t) = a \circ \Phi^t$. Then,

$$\frac{d}{dt} a_{\text{appr}}(t) = \{h, a_{\text{appr}}(t)\},$$

and we obtain Egorov's theorem,

$$e^{iHt/\varepsilon} \text{op}(a) e^{-iHt/\varepsilon} = \text{op}(a \circ \Phi^t) + O(\varepsilon^2).$$

2.1. The first correction

The asymptotic commutator expansion (11) allows to systematically derive higher order corrections to Egorov's theorem. For constructing the first correction, we set $a_{\text{appr}}(t) = a_0(t) + \varepsilon^2 a_2(t)$. The computation

$$\frac{i}{\varepsilon} [\text{op}(h), \text{op}(a_{\text{appr}}(t))] = \text{op}(\{h, a_{\text{appr}}(t)\}) - \frac{\varepsilon^2}{4} \text{op}(\{h, a_0(t)\}_3) + O(\varepsilon^4)$$

suggests to choose

$$a_0(t) = a \circ \Phi^t, \quad a_2(t) = -\frac{1}{4} \int_0^t \{h, a_0(\tau)\}_3 \circ \Phi^{t-\tau} d\tau.$$

Indeed, we compute the time derivative,

$$\begin{aligned} \frac{d}{dt} a_2(t) &= -\frac{1}{4} \left(\{h, a_0(t)\}_3 + \int_0^t \frac{d}{dt} (\{h, a_0(\tau)\}_3 \circ \Phi^{t-\tau}) d\tau \right) \\ &= -\frac{1}{4} \left(\{h, a_0(t)\}_3 + \int_0^t \{h, \{h, a_0(\tau)\}_3\} \circ \Phi^{t-\tau} d\tau \right) \\ &= -\frac{1}{4} \left(\{h, a_0(t)\}_3 + \{h, \int_0^t \{h, a_0(\tau)\}_3 \circ \Phi^{t-\tau} d\tau \} \right), \end{aligned}$$

where the last equation uses that the classical flow as a symplectic transformation of phase space preserves the Poisson bracket. We therefore obtain

$$\frac{d}{dt} a_{\text{appr}}(t) = \{h, a_{\text{appr}}(t)\} - \frac{\varepsilon^2}{4} \{h, a_0(t)\}_3.$$

Consequently,

$$e^{iHt/\varepsilon} \text{op}(a) e^{-iHt/\varepsilon} = \text{op}(a_{\text{appr}}(t)) + O(\varepsilon^4).$$

Remark 2.1. If $a : \mathbb{R}^{2d} \rightarrow \mathbb{R}$ is an observable invariant along the Hamiltonian flow, that is, $a \circ \Phi^t = a$ for all $t \in \mathbb{R}$, and additionally $\{h, a\}_3 = 0$, then the corrected approximation is also invariant,

$$a_{\text{appr}}(t) = a \circ \Phi^t - \frac{\varepsilon^2}{4} \int_0^t \{h, a \circ \Phi^\tau\}_3 \circ \Phi^{t-\tau} d\tau = a$$

for all $t \in \mathbb{R}$. In particular, mass ($a = 1$) and energy ($a = h$) are conserved.

2.2. Ordinary Differential Equations for the Correction

Our next aim is to reformulate the correction as

$$a_2(t) = -\frac{1}{4} \left((D^3 a \circ \Phi^t)_{ijk} \Lambda_{kji}^t + 3(D^2 a \circ \Phi^t)_{ij} \Gamma_{ji}^t + (Da \circ \Phi^t)_i \Xi_i^t \right),$$

where the components of the time-dependent tensors Λ^t , Γ^t , and Ξ^t satisfy a first order system of coupled ordinary differential equations, that is independent of the observable a and can be efficiently solved alongside the Hamiltonian flow Φ^t . Here and in the following, we use Einstein's summation convention for notational brevity.

Let

$$J = \begin{pmatrix} 0 & \text{Id} \\ -\text{Id} & 0 \end{pmatrix} \in \mathbb{R}^{2d \times 2d}.$$

We observe that we can write

$$\begin{aligned} a_2(t) &= -\frac{1}{4} \int_0^t \{h, a \circ \Phi^\tau\}_3 \circ \Phi^{t-\tau} d\tau \\ &= -\frac{1}{4} \int_0^t \left((D^3(a \circ \Phi^\tau))_{lmn} \left(JD^3h \right)_{nml} \right) \circ \Phi^{t-\tau} d\tau, \end{aligned}$$

where the 3-tensor (JD^3h) is defined by

$$\left(\widetilde{D^3h} \right)_{ijk} = \begin{cases} \frac{1}{6} (D^3h)_{ijk}, & i = j = k \\ \frac{1}{2} (D^3h)_{ijk}, & i = j \neq k \text{ or } i = k \neq j \text{ or } k = j \neq i \\ (D^3h)_{ijk}, & \text{else} \end{cases}$$

and

$$\left(JD^3h \right)_{ijk} = J_{il} J_{jm} J_{kn} \left(\widetilde{D^3h} \right)_{lmn}. \quad (12)$$

A closer look at this 3-tensor reveals that it is symmetric:

Lemma 2.1. *Let $k \in \mathbb{N}$ and $A = (a)_{i_1 \dots i_k} \in \mathbb{R}^{2d \times \dots \times 2d}$ be a k -tensor. Then,*

$$J_{i_1 l_1} \cdots J_{i_k l_k} A_{l_1 \dots l_k} \text{ is symmetric } \Leftrightarrow A \text{ is symmetric.}$$

In particular, the 3-tensor (JD^3h) defined in (12) is symmetric.

Proof. For every $m \in \{1, \dots, k\}$

$$J_{i_m l} A_{i_1 \dots i_{m-1} l i_{m+1} \dots i_k} = \begin{cases} A_{i_1 \dots i_{m-1} (i_m+d) i_{m+1} \dots i_k}, & i_m \leq d \\ -A_{i_1 \dots i_{m-1} (i_m-d) i_{m+1} \dots i_k}, & \text{else} \end{cases}.$$

Let $1 \leq j \leq k$. For $i_j \leq d$, we set $\alpha_j = 0$, $\beta_j = i_j + d$ and otherwise $\alpha_j = 1$, $\beta_j = i_j - d$. Then,

$$J_{i_1 l_1} \cdots J_{i_k l_k} A_{l_1 \dots l_k} = (-1)^{\alpha_1} \cdots (-1)^{\alpha_k} A_{\beta_1 \dots \beta_k}.$$

Hence, a permutation of (i_1, \dots, i_k) results in a permutation of $(\alpha_1, \dots, \alpha_k)$ and $(\beta_1, \dots, \beta_k)$. Thus, $J_{i_1 l_1} \cdots J_{i_k l_k} A_{l_1 \dots l_k}$ is symmetric if and only if A is symmetric. \square

We compute the third derivative of $a \circ \Phi^\tau$ by the chain rule, where we denote the derivatives of Φ^τ by $\partial_{i_k} \cdots \partial_{i_1} \Phi_j^\tau = (D^k \Phi^\tau)_{ji_1 \dots i_k}$,

$$\begin{aligned} (D^3(a \circ \Phi^\tau))_{lmn} &= (D^3 a \circ \Phi^\tau)_{ijk} (D\Phi^\tau)_{il} (D\Phi^\tau)_{jm} (D\Phi^\tau)_{kn} \\ &\quad + (D^2 a \circ \Phi^\tau)_{ij} (D^2 \Phi^\tau)_{ilm} (D\Phi^\tau)_{jn} + (D^2 a)_{ij} (D^2 \Phi^\tau)_{iln} (D\Phi^\tau)_{jm} \\ &\quad + (D^2 a \circ \Phi^\tau)_{ij} (D^2 \Phi^\tau)_{imn} (D\Phi^\tau)_{jl} + (Da \circ \Phi^\tau)_i (D^3 \Phi^\tau)_{ilmn}. \end{aligned}$$

Since the 3-tensor (JD^3h) is symmetric and $\Phi^\tau \circ \Phi^{t-\tau} = \Phi^t$, we obtain the reformulation

$$a_2(t) = -\frac{1}{4} \left((D^3 a \circ \Phi^t)_{ijk} \Lambda_{kji}^t + 3 (D^2 a \circ \Phi^t)_{ij} \Gamma_{ji}^t + (Da \circ \Phi^t)_i \Xi_i^t \right)$$

with

$$\begin{aligned}\Lambda_{ijk}^t &= \int_0^t \left[(D\Phi^\tau)_{il} (D\Phi^\tau)_{jm} (D\Phi^\tau)_{kn} \left(JD^3h \right)_{nml} \right] \circ \Phi^{t-\tau} d\tau, \\ \Gamma_{ij}^t &= \int_0^t \left[(D^2\Phi^\tau)_{ikl} (D\Phi^\tau)_{jm} \left(JD^3h \right)_{mlk} \right] \circ \Phi^{t-\tau} d\tau, \\ \Xi_i^t &= \int_0^t \left[(D^3\Phi^\tau)_{ijkl} \left(JD^3h \right)_{lkj} \right] \circ \Phi^{t-\tau} d\tau.\end{aligned}\tag{13}$$

Next, we compute the time derivatives of integrals, which are of the form observed in the defining equations of the time-dependent tensors Λ^t , Γ^t , and Ξ^t .

Lemma 2.2. *Let $b : \mathbb{R}^{2d} \rightarrow \mathbb{R}$ and $f : \mathbb{R} \times \mathbb{R}^{2d} \rightarrow \mathbb{R}$ smooth functions. Then,*

$$\frac{d}{dt} \int_0^t (bf(\tau)) \circ \Phi^{t-\tau} d\tau = \int_0^t \left(b \frac{d}{d\tau} f(\tau) \right) \circ \Phi^{t-\tau} d\tau + (bf(0)) \circ \Phi^t.$$

Proof. We start with

$$\begin{aligned}\frac{d}{dt} \int_0^t (bf(\tau)) \circ \Phi^{t-\tau} d\tau &= bf(t) + \int_0^t b \circ \Phi^{t-\tau} (Df(\tau))^T \circ \Phi^{t-\tau} \frac{d}{dt} \Phi^{t-\tau} d\tau \\ &\quad + \int_0^t (Db)^T \circ \Phi^{t-\tau} \frac{d}{dt} \Phi^{t-\tau} f(\tau) \circ \Phi^{t-\tau} d\tau.\end{aligned}$$

For introducing the τ -derivative in the integral, we compute

$$\begin{aligned}\frac{d}{d\tau} (bf(\tau) \circ \Phi^{t-\tau}) &= b \circ \Phi^{t-\tau} \left(\left(\frac{d}{d\tau} f(\tau) \right) \circ \Phi^{t-\tau} - (Df(\tau))^T \circ \Phi^{t-\tau} \frac{d}{dt} \Phi^{t-\tau} \right) \\ &\quad - (Db)^T \circ \Phi^{t-\tau} \frac{d}{dt} \Phi^{t-\tau} f(\tau) \circ \Phi^{t-\tau}.\end{aligned}$$

Therefore,

$$\begin{aligned}\frac{d}{dt} \int_0^t (bf(\tau)) \circ \Phi^{t-\tau} d\tau &= bf(t) + \int_0^t \left(b \frac{d}{d\tau} f(\tau) \right) \circ \Phi^{t-\tau} d\tau - \int_0^t \frac{d}{d\tau} ((bf(\tau)) \circ \Phi^{t-\tau}) d\tau \\ &= \int_0^t \left(b \frac{d}{d\tau} f(\tau) \right) \circ \Phi^{t-\tau} d\tau + (bf(0)) \circ \Phi^t.\end{aligned}$$

□

Now we are ready to formulate and prove our first main result, the explicit system of ordinary differential equations describing the second order correction to Egorov's theorem. In M. Zworski's recent monograph, the higher order terms are referred to "as difficult to compute", see [26, §11.1]. Our result shows nonetheless, that the computation is feasible.

Theorem 2.1 (Second Correction). *Let $h : \mathbb{R}^{2d} \rightarrow \mathbb{R}$ be a smooth function of subquadratic growth, $t \in \mathbb{R}$, and $\Phi^t : \mathbb{R}^{2d} \rightarrow \mathbb{R}^{2d}$ the Hamiltonian flow associated with h .*

Let $a : \mathbb{R}^{2d} \rightarrow \mathbb{R}$ be a Schwartz function. Then, there exists a constant $C = C(a, h, t) > 0$ such that for all $\varepsilon > 0$

$$\left\| e^{\text{iop}(h)t/\varepsilon} \text{op}(a) e^{-\text{iop}(h)t/\varepsilon} - \text{op}(a_0(t) + \varepsilon^2 a_2(t)) \right\| \leq C\varepsilon^4$$

where $a_0(t) = a \circ \Phi^t$ and

$$a_2(t) = -\frac{1}{4} \left((D^3 a \circ \Phi^t)_{ijk} \Lambda_{kji}^t + 3 (D^2 a \circ \Phi^t)_{ij} \Gamma_{ji}^t + (Da \circ \Phi^t)_i \Xi_i^t \right)$$

and the functions $\Lambda_{ijk}^t, \Gamma_{ij}^t, \Xi_i^t, i, j, k = 1, \dots, 2d$, solve the ordinary differential system

$$\begin{aligned} \frac{d}{dt} \Lambda_{ijk}^t &= M_{il}^t \Lambda_{ljk}^t + M_{jl}^t \Lambda_{ilk}^t + M_{kl}^t \Lambda_{ijl}^t + C_{ijk}^1(t), \\ \frac{d}{dt} \Gamma_{ij}^t &= (C_i^2(t))_{kl} \Lambda_{lkj}^t + M_{il}^t \Gamma_{lj}^t + M_{jl}^t \Gamma_{il}^t, \\ \frac{d}{dt} \Xi_i^t &= (C_i^3(t))_{jkl} \Lambda_{lkj}^t + 3 (C_i^2(t))_{jk} \Gamma_{kj}^t + M_{il}^t \Xi_l^t, \\ \Lambda_{ijk}^0 &= \Gamma_{ij}^0 = \Xi_i^0 = 0, \end{aligned} \tag{14}$$

with $M^t = J \cdot D^2 h \circ \Phi^t$ and

$$\begin{aligned} C_{ijk}^1(t) &= \left(J \widetilde{D^3 h} \circ \Phi^t \right)_{ijk}, & (C_i^2(t))_{jk} &= (J \cdot D^3 h \circ \Phi^t)_{ijk}, \\ (C_i^3(t))_{jkl} &= (J \cdot D^4 h \circ \Phi^t)_{ijkl}, \end{aligned}$$

where for a matrix $A \in \mathbb{R}^{2d \times 2d}$ and k -tensor $B \in \mathbb{R}^{2d \times \dots \times 2d}$, $A \cdot B \in \mathbb{R}^{2d \times \dots \times 2d}$ is given by $(A \cdot B)_{i_1 \dots i_k} = A_{i_1 s} B_{s i_2 \dots i_k}$ and $J \widetilde{D^3 h}$ is defined by (12).

Proof. The initial values $\Lambda_{ijk}^0 = \Gamma_{ij}^0 = \Xi_i^0 = 0$ are clear, since $\Lambda_{ijk}^t, \Gamma_{ij}^t$ and Ξ_i^t are defined in (13) as an integral from 0 to t , over a smooth integrand. Next, we study the time derivative of Λ_{ijk}^t . For this, we write the Hamiltonian system as

$$\frac{d}{dt} \Phi^t = J D h \circ \Phi^t, \tag{15}$$

and apply the differential operator D to obtain the Jacobi stability equation

$$\frac{d}{dt} (D \Phi^t)_{ij} = (J \cdot D^2 h \circ \Phi^t)_{im} (D \Phi^t)_{mj}.$$

Then, by Lemma 2.2,

$$\begin{aligned} \frac{d}{dt} \Lambda_{ijk}^t &= \int_0^t \left[\frac{d}{d\tau} \left((D \Phi^\tau)_{il} (D \Phi^\tau)_{jm} (D \Phi^\tau)_{kn} \right) \left(J \widetilde{D^3 h} \right)_{lmn} \right] \circ \Phi^{t-\tau} d\tau \\ &\quad + \left[(D \Phi^0)_{il} (D \Phi^0)_{jm} (D \Phi^0)_{kn} \left(J \widetilde{D^3 h} \right)_{lmn} \right] \circ \Phi^t \\ &= \int_0^t \left[(J \cdot D^2 h \circ \Phi^\tau)_{i\nu} (D \Phi^\tau)_{\nu l} (D \Phi^\tau)_{jm} (D \Phi^\tau)_{kn} \left(J \widetilde{D^3 h} \right)_{lmn} \right] \circ \Phi^{t-\tau} d\tau \\ &\quad + \int_0^t \left[(D \Phi^\tau)_{il} (J \cdot D^2 h \circ \Phi^\tau)_{j\nu} (D \Phi^\tau)_{\nu m} (D \Phi^\tau)_{kn} \left(J \widetilde{D^3 h} \right)_{lmn} \right] \circ \Phi^{t-\tau} d\tau \\ &\quad + \int_0^t \left[(D \Phi^\tau)_{il} (D \Phi^\tau)_{jm} (J \cdot D^2 h \circ \Phi^\tau)_{k\nu} (D \Phi^\tau)_{\nu n} \left(J \widetilde{D^3 h} \right)_{lmn} \right] \circ \Phi^{t-\tau} d\tau \\ &\quad + \left(J \widetilde{D^3 h} \right)_{ijk} \circ \Phi^t. \end{aligned}$$

So we can finish the proof for $\frac{d}{dt}\Lambda^t$ by using that $\Phi^\tau \circ \Phi^{t-\tau} = \Phi^t$. The proofs for $\frac{d}{dt}\Gamma^t$ and $\frac{d}{dt}\Xi^t$ are analogous: We differentiate the Jacobi stability equation to obtain

$$\begin{aligned}\frac{d}{dt}(D^2\Phi^t)_{ijk} &= (J \cdot D^3h \circ \Phi^t)_{imn} (D\Phi^t)_{nk} (D\Phi^t)_{mj} + (J \cdot D^2h \circ \Phi^t)_{im} (D^2\Phi^t)_{mjk}, \\ \frac{d}{dt}(D^3\Phi^t)_{ijkl} &= (J \cdot D^4h \circ \Phi^t)_{imn\mu} (D\Phi^t)_{nk} (D\Phi^t)_{mj} (D\Phi^t)_{\mu l} \\ &\quad + (J \cdot D^3h \circ \Phi^t)_{imn} (D\Phi^t)_{mj} (D^2\Phi^t)_{nkl} \\ &\quad + (J \cdot D^3h \circ \Phi^t)_{imn} (D\Phi^t)_{nk} (D^2\Phi^t)_{mjl} \\ &\quad + (J \cdot D^3h \circ \Phi^t)_{imn} (D\Phi^t)_{nl} (D^2\Phi^t)_{mjk} + (J \cdot D^2h \circ \Phi^t)_{im} (D^3\Phi^t)_{mjkl}.\end{aligned}$$

Moreover, we use

$$\begin{aligned}\frac{d}{d\tau} \left((D^2\Phi^\tau)_{ikl} (D\Phi^\tau)_{jm} \right) \left(J\widetilde{D^3h} \right)_{mlk} \\ = \left[(J \cdot D^3h \circ \Phi^\tau)_{i\mu\nu} (D\Phi^\tau)_{\nu l} (D\Phi^\tau)_{\mu k} (D\Phi^\tau)_{jm} \right. \\ \left. + (J \cdot D^2h \circ \Phi^\tau)_{i\mu} (D^2\Phi)_{\mu kl} (D\Phi^\tau)_{jm} \right. \\ \left. + (D^2\Phi^\tau)_{ikl} (J \cdot D^2h \circ \Phi^\tau)_{j\mu} (D\Phi^\tau)_{\mu m} \right] \left(J\widetilde{D^3h} \right)_{mlk}, \\ \frac{d}{d\tau} (D^3\Phi^\tau)_{ijkl} \left(J\widetilde{D^3h} \right)_{lkj} = \left[(J \cdot D^4h \circ \Phi^\tau)_{i\mu\nu\eta} (D\Phi^\tau)_{\nu k} (D\Phi^\tau)_{\mu j} (D\Phi^\tau)_{\eta l} \right. \\ \left. + 3(J \cdot D^3h \circ \Phi^\tau)_{i\mu\nu} (D\Phi^\tau)_{\nu k} (D^2\Phi^\tau)_{\mu jl} \right. \\ \left. + (J \cdot D^2h \circ \Phi^\tau)_{i\mu} (D^3\Phi^\tau)_{\mu jkl} \right] \left(J\widetilde{D^3h} \right)_{lkj}.\end{aligned}$$

as well as $(J \cdot D^3h \circ \Phi^\tau)_{i\mu\nu} = (J \cdot D^3h \circ \Phi^\tau)_{i\nu\mu}$ and $D^2\Phi^0 = D^3\Phi^0 = 0$. \square

2.3. Vectorization: General Hamiltonians

For the numerical simulation of the ordinary differential system (14), we vectorize the tensors. Recall, that for matrices $A \in \mathbb{R}^{n \times n}$ and $B \in \mathbb{R}^{m \times m}$, the Kronecker product $A \otimes B \in \mathbb{R}^{mn \times mn}$ is defined as

$$A \otimes B = \begin{pmatrix} A_{11}B & \cdots & A_{1n}B \\ \vdots & & \vdots \\ A_{n1}B & \cdots & A_{nn}B \end{pmatrix}.$$

That is,

$$(A \otimes B)_{ij} = A_{i_1j_1} B_{i_2j_2}, \quad \text{if } i = (i_1 - 1)m + i_2, j = (j_1 - 1)m + j_2.$$

The vectorization of a k-tensor $C = (C)_{i_1 \dots i_k} \in \mathbb{R}^{n_1 \times \dots \times n_k}$ is defined as

$$\text{vec}(C) = \begin{pmatrix} c_{1 \dots 1} & c_{1 \dots 12} & \cdots & c_{1 \dots 1n_k} & c_{1 \dots 121} & \cdots & c_{n_1 \dots n_k} \end{pmatrix}^T \in \mathbb{R}^{n_1 \dots n_k}.$$

That is, $\text{vec}(C)_i = c_{i_1 \dots i_k}$ if

$$i = (i_1 - 1) \prod_{l=2}^k n_l + (i_2 - 1) \prod_{l=3}^k n_l + \cdots + (i_{k-1} - 1)n_k + i_k.$$

The following observation allows the vectorization of the products occuring on the right hand side of our differential equation (14).

Lemma 2.3. *Let $A \in \mathbb{R}^{m \times m}$ a matrix, $B \in \mathbb{R}^{m \times \dots \times m}$ an n -tensor, and $k \in \{1, \dots, m\}$. Define the n -tensor $C \in \mathbb{R}^{m \times \dots \times m}$ as*

$$C_{i_1 \dots i_n} = A_{i_k l} B_{i_1 \dots i_{k-1} l i_{k+1} \dots i_n}.$$

Then,

$$\text{vec}(C) = \left(\overbrace{\text{Id}_m \otimes \dots \otimes \text{Id}_m}^{k-1 \text{ times}} \otimes A \otimes \overbrace{\text{Id}_m \otimes \dots \otimes \text{Id}_m}^{n-k \text{ times}} \right) \cdot \text{vec}(B).$$

Proof. We define the matrix $\tilde{A} = \left(\overbrace{\text{Id}_m \otimes \dots \otimes \text{Id}_m}^{k-1 \text{ times}} \otimes A \otimes \overbrace{\text{Id}_m \otimes \dots \otimes \text{Id}_m}^{n-k \text{ times}} \right)$. That is,

$$\tilde{A}_{il} = \begin{cases} A_{i_k l_k}, & \text{if } i_\mu = l_\mu \text{ for all } \mu \in \{1, \dots, k-1, k+1, \dots, n\} \\ 0, & \text{else} \end{cases}$$

for $i = (i_1 - 1)m^{n-1} + \dots + (i_{n-1} - 1)m + i_n$ and $l = (l_1 - 1)m^{n-1} + \dots + (l_{n-1} - 1)m + l_n$. Then,

$$\text{vec}(C)_i = \tilde{A}_{il} \cdot \text{vec}(B)_l = A_{i_k l_k} B_{i_1 \dots i_{k-1} l_k i_{k+1} \dots i_n} = C_{i_1 \dots i_n}$$

for $i = (i_1 - 1)m^{n-1} + \dots + (i_{n-1} - 1)m + i_n$. \square

Now we reformulate the results of Theorem 2.1 in vectorized form.

Corollary 2.1 (Vectorization). *Consider the time-dependent tensors M^t and $C^j(t)$, $j = 1, 2, 3$, together with the Hamiltonian flow Φ^t of Theorem 2.1. Let the functions Λ_{ijk}^t , Γ_{ij}^t and Ξ_i^t solve the ordinary differential system*

$$\begin{aligned} \frac{d}{dt} \Lambda_{ijk}^t &= M_{il}^t \Lambda_{ljk}^t + M_{jl}^t \Lambda_{ilk}^t + M_{kl}^t \Lambda_{ijl}^t + C_{ijk}^1(t), \\ \frac{d}{dt} \Gamma_{ij}^t &= (C_i^2(t))_{kl} \Lambda_{lkj}^t + M_{il}^t \Gamma_{lj}^t + M_{jl}^t \Gamma_{il}^t, \\ \frac{d}{dt} \Xi_i^t &= (C_i^3(t))_{jkl} \Lambda_{lkj}^t + 3 (C_i^2(t))_{jk} \Gamma_{kj}^t + M_{il}^t \Xi_l^t. \end{aligned}$$

Then,

$$\frac{d}{dt} \begin{pmatrix} \Phi^t \\ \text{vec}(\Lambda^t) \\ \text{vec}(\Gamma^t) \\ \Xi^t \end{pmatrix} = \begin{pmatrix} \text{Id}_{2d} & 0 & 0 & 0 \\ 0 & K^t & 0 & 0 \\ 0 & D^t & L^t & 0 \\ 0 & C_m^3(t) & 3C_m^2(t) & M^t \end{pmatrix} \begin{pmatrix} J \cdot Dh \circ \Phi^t \\ \text{vec}(\Lambda^t) \\ \text{vec}(\Gamma^t) \\ \Xi^t \end{pmatrix} + \begin{pmatrix} 0 \\ C_v^1(t) \\ 0 \\ 0 \end{pmatrix} \quad (16)$$

with $C_v^1(t) = \text{vec}(C^1(t))$ and

$$C_m^j(t) = \begin{pmatrix} \text{vec}(C_1^j(t)) & \dots & \text{vec}(C_{2d}^j(t)) \end{pmatrix}^T, \quad j = 2, 3,$$

and

$$\begin{aligned} K^t &= M^t \otimes \text{Id}_{2d} \otimes \text{Id}_{2d} + \text{Id}_{2d} \otimes M^t \otimes \text{Id}_{2d} + \text{Id}_{2d} \otimes \text{Id}_{2d} \otimes M^t, \\ D^t &= \text{Id}_{2d} \otimes C_m^2(t), \quad L^t = M^t \otimes \text{Id}_{2d} + \text{Id}_{2d} \otimes M^t. \end{aligned}$$

Proof. Applying Lemma 2.3 to the Λ_{ijk}^t part of the ordinary differential equation, we obtain

$$\begin{aligned} \frac{d}{dt} \vec{\Lambda}^t &= \frac{d}{dt} \text{vec}(\Lambda_{ijk}^t) = \text{vec}(M_{il}^t \Lambda_{ljk}^t + M_{jl}^t \Lambda_{ilk}^t + M_{kl}^t \Lambda_{ijl}^t + C_{ijk}^1(t)) \\ &= (M^t \otimes \text{Id}_{2d} \otimes \text{Id}_{2d} + \text{Id}_{2d} \otimes M^t \otimes \text{Id}_{2d} + \text{Id}_{2d} \otimes \text{Id}_{2d} \otimes M^t) \text{vec}(\Lambda^t) + C_v^1(t) \\ &= K^t \text{vec}(\Lambda^t) + C_v^1(t). \end{aligned}$$

The proofs for $\text{vec}(\Gamma^t)$ and Ξ^t are analogous. \square

2.4. Vectorization: Schrödinger Hamiltonians

We now analyse the vectorized system of ordinary differential equations (16) for the special case, that the Hamilton function is given by

$$h(q, p) = \frac{1}{2}|p|^2 + V(q).$$

Then,

$$D^2 h = \begin{pmatrix} D^2 V & 0 \\ 0 & \text{Id}_d \end{pmatrix}, \quad M^t = J \cdot D^2 h \circ \Phi^t = \begin{pmatrix} 0 & \text{Id}_d \\ -D^2 V \circ \Phi_q^t & 0 \end{pmatrix}. \quad (17)$$

Moreover,

$$D^3 h = \begin{cases} (D^3 V)_{ijk}, & i, j, k \leq d \\ 0, & \text{else} \end{cases}, \quad D^4 h = \begin{cases} (D^4 V)_{ijkl}, & i, j, k, l \leq d \\ 0, & \text{else} \end{cases}.$$

To exploit this zero pattern, we reorder our system of differential equations. We set

$$\begin{aligned} (\Lambda_1^t)_{ijk} &= \Lambda_{ijk}^t, & (\Lambda_{2,1}^t)_{ijk} &= \Lambda_{(i+d)jk}^t, & (\Lambda_{2,2}^t)_{ijk} &= \Lambda_{i(j+d)k}^t, \\ (\Lambda_{2,3}^t)_{ijk} &= \Lambda_{ij(k+d)}^t, & (\Lambda_{3,1}^t)_{ijk} &= \Lambda_{i(j+d)(k+d)}^t, & (\Lambda_{3,2}^t)_{ijk} &= \Lambda_{(i+d)j(k+d)}^t, \\ (\Lambda_{3,3}^t)_{ijk} &= \Lambda_{(i+d)(j+d)k}^t, & (\Lambda_4^t)_{ijk} &= \Lambda_{(i+d)(j+d)(k+d)}^t, \\ (\Gamma_1^t)_{ij} &= \Gamma_{ij}^t, & (\Gamma_{2,1}^t)_{ij} &= \Gamma_{(i+d)j}^t, & (\Gamma_{2,2}^t)_{ij} &= \Gamma_{i(j+d)}^t, \\ (\Gamma_3^t)_{ij} &= \Gamma_{(i+d)(j+d)}^t, & (\Xi_1^t)_i &= \Xi_i^t, & (\Xi_2^t)_i &= \Xi_{i+d}^t, \end{aligned}$$

for $i, j, k = 1, \dots, d$. These terms are recollected according to

$$\vec{\Lambda}_1^t = \text{vec}(\Lambda_1^t), \quad \vec{\Lambda}_4^t = \text{vec}(\Lambda_4^t), \quad \vec{\Gamma}_1^t = \text{vec}(\Gamma_1^t), \quad \vec{\Gamma}_3^t = \text{vec}(\Gamma_3^t),$$

and

$$\vec{\Lambda}_2^t = \begin{pmatrix} \text{vec}(\Lambda_{2,1}^t) \\ \text{vec}(\Lambda_{2,2}^t) \\ \text{vec}(\Lambda_{2,3}^t) \end{pmatrix}, \quad \vec{\Lambda}_3^t = \begin{pmatrix} \text{vec}(\Lambda_{3,1}^t) \\ \text{vec}(\Lambda_{3,2}^t) \\ \text{vec}(\Lambda_{3,3}^t) \end{pmatrix}, \quad \vec{\Gamma}_2^t = \begin{pmatrix} \text{vec}(\Gamma_{2,1}^t) \\ \text{vec}(\Gamma_{2,2}^t) \end{pmatrix}.$$

This reordering enhances the zero pattern of the right hand side of the ordinary differential equation.

Theorem 2.2 (Schrödinger Hamiltonians). *Let $V : \mathbb{R}^d \rightarrow \mathbb{R}$ be a smooth function of subquadratic growth and $h : \mathbb{R}^{2d} \rightarrow \mathbb{R}$, $h(q, p) = \frac{1}{2}|p|^2 + V(q)$. Let $(q_0, p_0) \in \mathbb{R}^{2d}$ and*

$(\Phi^t, \text{vec}(\Lambda^t), \text{vec}(\Gamma^t), \Xi^t)$ be the solution of the ordinary differential equation (16) given in Corollary 2.1 with initial values

$$(\Phi^0, \text{vec}(\Lambda^0), \text{vec}(\Gamma^0), \Xi^0) = ((q_0, p_0), 0, 0, 0) \in \mathbb{R}^{2d+8d^3+4d^2+2d}.$$

We set

$$\Psi_1^t = \Phi_q^t \in \mathbb{R}^d, \quad \Psi_2^t = \begin{pmatrix} \Phi_p^t \\ \vec{\Lambda}_2^t \\ \vec{\Lambda}_4^t \\ \vec{\Gamma}_2^t \\ \Xi_2^t \end{pmatrix} \in \mathbb{R}^{4d^3+2d^2+2d}, \quad \Psi_3^t = \begin{pmatrix} \vec{\Lambda}_1^t \\ \vec{\Lambda}_3^t \\ \vec{\Gamma}_1^t \\ \vec{\Gamma}_3^t \\ \Xi_1^t \end{pmatrix} \in \mathbb{R}^{4d^3+2d^2+d}.$$

Then,

$$\frac{d}{dt} \begin{pmatrix} \Psi_1^t \\ \Psi_2^t \\ \Psi_3^t \end{pmatrix} = \begin{pmatrix} 0 & A_1 & 0 \\ 0 & 0 & A_2 \circ \Psi_1^t \\ 0 & A_3 \circ \Psi_1^t & 0 \end{pmatrix} \cdot \begin{pmatrix} \Psi_1^t \\ \Psi_2^t \\ \Psi_3^t \end{pmatrix} + \begin{pmatrix} 0 \\ b_2 \circ \Psi_1^t \\ 0 \end{pmatrix} \quad (18)$$

with $\Psi_1^0 = q_0$, $\Psi_2^0 = (p_0, 0)$ and $\Psi_3^0 = 0$, where $A_1 = \begin{pmatrix} \text{Id}_d & 0 & 0 & 0 & 0 \end{pmatrix}$ and

$$A_2 = \begin{pmatrix} 0 & 0 & 0 & 0 & 0 \\ K_2 & K_3 & 0 & 0 & 0 \\ 0 & K_6 & 0 & 0 & 0 \\ K_8 & 0 & K_9 & K_{15} & 0 \\ K_{12} & 0 & K_{13} & 0 & K_{14} \end{pmatrix}, \quad b_2 = \begin{pmatrix} -DV \\ 0 \\ -\text{vec}(\widetilde{D^3V}) \\ 0 \\ 0 \end{pmatrix},$$

and

$$A_3 = \begin{pmatrix} 0 & K_1 & 0 & 0 & 0 \\ 0 & K_4 & K_5 & 0 & 0 \\ 0 & 0 & 0 & K_7 & 0 \\ 0 & K_{10} & 0 & K_{11} & 0 \\ 0 & 0 & 0 & 0 & \text{Id}_d \end{pmatrix}.$$

Denoting

$$(D^3V)_m = \begin{pmatrix} D^3V_{(1,1,1)} & \cdots & D^3V_{(1,1,d)} & D^3V_{(1,2,1)} & \cdots & D^3V_{(1,d,d)} \\ \vdots & \vdots & \vdots & \vdots & \vdots & \vdots \\ D^3V_{(d,1,1)} & \cdots & D^3V_{(d,1,d)} & D^3V_{(d,2,1)} & \cdots & D^3V_{(d,d,d)} \end{pmatrix}$$

and

$$(D^4V)_m = \begin{pmatrix} D^4V_{(1,1,1,1)} & \cdots & D^4V_{(1,1,1,d)} & D^4V_{(1,1,2,1)} & \cdots & D^4V_{(1,1,d,d)} & D^4V_{(1,2,1,1)} & \cdots & D^4V_{(1,d,d,d)} \\ \vdots & \vdots & \vdots & \vdots & \vdots & \vdots & \vdots & \vdots & \vdots \\ D^4V_{(d,1,1,1)} & \cdots & D^4V_{(d,1,1,d)} & D^4V_{(d,1,2,1)} & \cdots & D^4V_{(d,1,d,d)} & D^4V_{(d,2,1,1)} & \cdots & D^4V_{(d,d,d,d)} \end{pmatrix},$$

the matrices K_1, \dots, K_{15} are given by

$$\begin{aligned}
 K_1 &= \begin{pmatrix} \text{Id}_{d^3} & \text{Id}_{d^3} & \text{Id}_{d^3} \end{pmatrix}, K_2 = \begin{pmatrix} -D^2V \otimes \text{Id}_d \otimes \text{Id}_d \\ \text{Id}_d \otimes -D^2V \otimes \text{Id}_d \\ \text{Id}_d \otimes \text{Id}_d \otimes -D^2V \end{pmatrix}, \\
 K_3 &= \begin{pmatrix} 0 & \text{Id}_{d^3} & \text{Id}_{d^3} \\ \text{Id}_{d^3} & 0 & \text{Id}_{d^3} \\ \text{Id}_{d^3} & \text{Id}_{d^3} & 0 \end{pmatrix}, \\
 K_4 &= \begin{pmatrix} 0 & \text{Id}_d \otimes \text{Id}_d \otimes -D^2V & \text{Id}_d \otimes -D^2V \otimes \text{Id}_d \\ \text{Id}_d \otimes \text{Id}_d \otimes -D^2V & 0 & -D^2V \otimes \text{Id}_d \otimes \text{Id}_d \\ \text{Id}_d \otimes -D^2V \otimes \text{Id}_d & -D^2V \otimes \text{Id}_d \otimes \text{Id}_d & 0 \end{pmatrix}, \\
 K_5 &= \begin{pmatrix} \text{Id}_{d^3} \\ \text{Id}_{d^3} \\ \text{Id}_{d^3} \end{pmatrix}, \\
 K_6 &= \begin{pmatrix} -D^2V \otimes \text{Id}_d \otimes \text{Id}_d & \text{Id}_d \otimes -D^2V \otimes \text{Id}_d & \text{Id}_d \otimes \text{Id}_d \otimes -D^2V \end{pmatrix}, \\
 K_7 &= \begin{pmatrix} \text{Id}_{d^2} & \text{Id}_{d^2} \end{pmatrix}, K_8 = \begin{pmatrix} 0 \\ -(D^3V)_m \otimes \text{Id}_d \end{pmatrix}, K_9 = \begin{pmatrix} \text{Id}_d \otimes -D^2V \\ -D^2V \otimes \text{Id}_d \end{pmatrix}, \\
 K_{10} &= \begin{pmatrix} 0 & 0 & -(D^3V)_m \otimes \text{Id}_d \end{pmatrix}, K_{11} = \begin{pmatrix} -D^2V \otimes \text{Id}_d & \text{Id}_d \otimes -D^2V \end{pmatrix}, \\
 K_{12} &= -(D^4V)_m, K_{13} = -3(D^3V)_m, K_{14} = -(D^3V)_m, K_{15} = \begin{pmatrix} \text{Id}_{d^2} \\ \text{Id}_{d^2} \end{pmatrix}.
 \end{aligned}$$

Proof. In the following, we denote

$$A_{i_1, \dots, s, \dots, i_k} B_{j_1, \dots, s, \dots, j_l} = \sum_{s=1}^d A_{i_1, \dots, s, \dots, i_k} B_{j_1, \dots, s, \dots, j_l}$$

for tensors $A \in \mathbb{R}^{d \times \dots \times d}$ and $B \in \mathbb{R}^{2d \times \dots \times 2d}$. By the specific form of the matrix M^t given in (17), we obtain

$$\begin{aligned}
 M_{il_1}^t \Lambda_{l_1 l_2 l_3}^t &= \begin{cases} \Lambda_{(i+d)l_2 l_3}^t, & \text{for } i \leq d, \\ (-D^2V \circ \Phi_q^t)_{(i-d)s} \cdot \Lambda_{sl_2 l_3}^t, & \text{else,} \end{cases} \\
 M_{il_2}^t \Lambda_{l_1 l_2 l_3}^t &= \begin{cases} \Lambda_{l_1 (i+d)l_3}^t, & \text{for } i \leq d, \\ (-D^2V \circ \Phi_q^t)_{(i-d)s} \cdot \Lambda_{l_1 sl_3}^t, & \text{else,} \end{cases} \\
 M_{il_3}^t \Lambda_{l_1 l_2 l_3}^t &= \begin{cases} \Lambda_{l_1 l_2 (i+d)}^t, & \text{for } i \leq d, \\ (-D^2V \circ \Phi_q^t)_{(i-d)s} \cdot \Lambda_{l_1 l_2 s}^t, & \text{else,} \end{cases}
 \end{aligned}$$

Therefore,

$$\begin{aligned}
 \frac{d}{dt} \Lambda_{ijk}^t &= 1_{\{i \leq d\}} \Lambda_{(i+d)jk}^t - 1_{\{i > d\}} (D^2V \circ \Phi_q^t)_{(i-d)s} \cdot \Lambda_{sjk}^t \\
 &\quad + 1_{\{j \leq d\}} \Lambda_{i(j+d)k}^t - 1_{\{j > d\}} (D^2V \circ \Phi_q^t)_{(j-d)s} \cdot \Lambda_{isk}^t \\
 &\quad + 1_{\{k \leq d\}} \Lambda_{ij(k+d)}^t - 1_{\{k > d\}} (D^2V \circ \Phi_q^t)_{(k-d)s} \cdot \Lambda_{ijs}^t
 \end{aligned}$$

$$- 1_{\{i,j,k>d\}} \widetilde{D^3V}_{(i-d)(j-d)(k-d)} \circ \Phi_q^t,$$

since

$$C_{ijk}^1(t) = \begin{cases} -\widetilde{D^3V}_{(i-d)(j-d)(k-d)} \circ \Phi_q^t, & i, j, k > d \\ 0, & \text{else} \end{cases}.$$

In the same way, we obtain

$$\begin{aligned} \frac{d}{dt} \Gamma_{ij}^t &= 1_{\{i \leq d\}} \Gamma_{(i+d)j}^t - 1_{\{i > d\}} (D^2V \circ \Phi_q^t)_{(i-d)s} \cdot \Gamma_{sj}^t \\ &\quad + 1_{\{j \leq d\}} \Gamma_{i(j+d)}^t - 1_{\{j > d\}} (D^2V \circ \Phi_q^t)_{(j-d)s} \cdot \Gamma_{is}^t \\ &\quad - 1_{\{i \leq d\}} (D^3V \circ \Phi_q^t)_{(i-d)ss'} \Lambda_{s'sj}, \end{aligned}$$

since

$$(C_i^2(t))_{jk} = \begin{cases} -(D^3V)_{(i-d)jk} \circ \Phi_q^t, & i > d, j, k \leq d \\ 0, & \text{else} \end{cases}.$$

Finally, we compute

$$\begin{aligned} \frac{d}{dt} \Xi_i^t &= 1_{\{i \leq d\}} \Xi_{(i+d)}^t - 1_{\{i > d\}} (D^2V \circ \Phi_q^t)_{(i-d)s} \cdot \Xi_s^t - 1_{\{i > d\}} (D^4V \circ \Phi_q^t)_{iss's''} \Lambda_{s''s's}^t \\ &\quad - 3 \cdot 1_{\{i > d\}} (D^3V \circ \Phi_q^t)_{iss'} \Gamma_{s's}^t, \end{aligned}$$

using that

$$(C_i^3(t))_{jkl} = \begin{cases} -(D^4V)_{(i-d)jkl} \circ \Phi_q^t, & i > d, j, k, l \leq d \\ 0, & \text{else} \end{cases}.$$

Reordering the above differential equations, we obtain the claimed result. \square

3. Discretization

We adopt the following general scheme which has been previously developed for discretizing Egorov's theorem [10]: The classical Hamiltonian flow Φ^t is discretized by a symplectic order p method $z_{n+1} = \Psi^\tau(z_n)$ with sufficiently small step size $\tau > 0$. The initial Wigner function is split into its positive and negative part $W_{\psi_0} = W_{\psi_0}^+ - W_{\psi_0}^-$ and is sampled by sufficiently many phase space points $z_1^\pm, \dots, z_N^\pm \in \mathbb{R}^{2d}$. Then, expectation values for various observables $a : \mathbb{R}^{2d} \rightarrow \mathbb{R}$ with respect to the solution ψ_t of the Schrödinger equation

$$i\varepsilon \partial \psi_t = H \psi_t$$

are approximated according to

$$\begin{aligned}
\langle \psi_t, \text{op}(a) \psi_t \rangle &= \langle \psi_0, e^{iHt/\varepsilon} \text{op}(a) e^{-iHt/\varepsilon} \psi_0 \rangle \\
&= \langle \psi_0, \text{op}(a \circ \Phi^t) \psi_0 \rangle + O(\varepsilon^2) = \int_{\mathbb{R}^{2d}} a(\Phi^t(z)) W_{\psi_0} dz + O(\varepsilon^2) \\
&\approx \frac{1}{N} \sum_{j=1}^N a((\Psi^\tau \circ \dots \circ \Psi^\tau)(z_j^+)) - \frac{1}{N} \sum_{j=1}^N a((\Psi^\tau \circ \dots \circ \Psi^\tau)(z_j^-)) \\
&=: I^N(a \circ \Psi^\tau \circ \dots \circ \Psi^\tau).
\end{aligned} \tag{19}$$

The computational work of this algorithm lies in the sampling of the initial Wigner function and the classical evolution of the sample points. Then, expectation values are computed by a final phase space summation.

Remark 3.1. *If the initial wave function ψ_0 is a Gaussian wave packet, then the Wigner function W_{ψ_0} is positive, see [6, Theorem 1.102], and the approximation (19) reads as*

$$\langle \psi_t, \text{op}(a) \psi_t \rangle \approx \frac{1}{N} \sum_{j=1}^N a((\Psi^\tau \circ \dots \circ \Psi^\tau)(z_j))$$

with $z_1, \dots, z_N \in \mathbb{R}^{2d}$ sampled according to W_{ψ_0} . If ψ_0 is a superposition of Gaussian wave packets, then the Wigner function W_{ψ_0} is a sum of phase space Gaussians plus oscillatory cross terms, such that stratified sampling can be applied, see [10, §3].

3.1. Splitting the two integrals

Here, we add the second order correction $\varepsilon^2 a_2(t)$ to $a_0(t) = a \circ \Phi^t$ and discretize

$$\langle \text{op}(a_0(t) + \varepsilon^2 a_2(t)) \psi_0, \psi_0 \rangle = \int_{\mathbb{R}^{2d}} a_0(t, z) W_{\psi_0}(z) dz + \varepsilon^2 \int_{\mathbb{R}^{2d}} a_2(t, z) W_{\psi_0}(z) dz,$$

where we split the phase space integral into two parts. This splitting has an impact on the computing time, since the prefactor ε^2 allows to discretize the second summand rather coarsely without diminishing the overall accuracy of the approximation. We approximate the two integrals via

$$\int_{\mathbb{R}^{2d}} a_j(t, z) W_{\psi_0}(z) dz \approx I^{N_j}(a_j(t)), \quad j = 0, 2,$$

such that $\langle \text{op}(a_0(t) + \varepsilon^2 a_2(t)) \psi_0, \psi_0 \rangle \approx I^{N_0}(a_0(t)) + \varepsilon^2 I^{N_2}(a_2(t))$.

3.2. Computing the integrals

The two integrals depend on the Wigner function of the initial wave function. Here, we consider a Gaussian wave packet centered at $(q_0, p_0) \in \mathbb{R}^{2d}$,

$$\psi_0(q) = (\pi\varepsilon)^{-d/4} \exp\left(- (2\varepsilon)^{-1} |q - q_0|^2 + \frac{i}{\varepsilon} p_0 \cdot (q - q_0)\right). \tag{20}$$

In this case the Wigner function can be calculated analytically as a phase space Gaussian with mean (q_0, p_0) and covariance matrix Id_{2d} (see, for example [10, §3]), namely

$$W_{\psi_0}(z) = (\pi\epsilon)^{-d} \exp\left(-\frac{1}{\epsilon}|z - (q_0, p_0)|^2\right).$$

Let $f : \mathbb{R} \times \mathbb{R}^{2d} \rightarrow \mathbb{R}$ be the function to be integrated. Due to the high dimensionality of the problem, we use quasi-Monte Carlo quadrature. That is,

$$\int_{\mathbb{R}^{2d}} f(t, z) W_{\psi_0}(z) dz \approx I^N(f(t)) = \frac{1}{N} \sum_{j=1}^N f(t, z_j),$$

with quadrature nodes $\{z_j\}_{j=1}^N \subset \mathbb{R}^{2d}$ of low star discrepancy with respect to the multivariate normal distribution. Then, the Koksma-Hlawka inequality yields a constant $\gamma = \gamma(f(t)) > 0$ such that

$$\left| \int_{\mathbb{R}^{2d}} f(t, z) W_{\psi_0}(z) dz - I^N(f(t)) \right| \leq \gamma (\log N)^{c_d} N^{-1}, \quad (21)$$

where $c_d \geq 2d$, see, for example, [10, §3.2].

3.3. Splitting the ordinary differential equations

To compute $a_0(t) = a \circ \Phi^t$ we have to discretize the Hamiltonian equation

$$\frac{d}{dt} \begin{pmatrix} \Phi_q^t \\ \Phi_p^t \end{pmatrix} = \begin{pmatrix} 0 & \text{Id}_d \\ 0 & 0 \end{pmatrix} \begin{pmatrix} \Phi_q^t \\ \Phi_p^t \end{pmatrix} + \begin{pmatrix} 0 \\ DV \circ \Phi_q^t \end{pmatrix}.$$

Now let ϕ_1^t and ϕ_2^t be the flows of the following differential equations

$$\frac{d}{dt} y(t) = \begin{pmatrix} 0 & \text{Id}_d \\ 0 & 0 \end{pmatrix} \cdot y(t), \quad \frac{d}{dt} y(t) = \begin{pmatrix} 0 \\ DV(y_1(t)) \end{pmatrix}.$$

These flows can be computed exactly by

$$\phi_1^t(y) = y + t \begin{pmatrix} y_2 \\ 0 \end{pmatrix}, \quad \phi_2^t = y + t \begin{pmatrix} 0 \\ DV(y_1) \end{pmatrix},$$

and

$$\phi^\tau = \phi_1^{\tau/2} \circ \phi_2^\tau \circ \phi_1^{\tau/2}$$

defines a symplectic second order splitting scheme for $\tau > 0$, the so-called Strang splitting, see [28, §4.3]. By suitable compositions of this scheme, one can construct symplectic splitting schemes of arbitrary order, see e.g. [27, §4]).

For computing the correction term $a_2(t)$, we write the ordinary differential equation (18) of Theorem 2.2 as

$$\frac{d}{dt} \Psi^t = N_1 \Psi^t + ((N_2 \circ \Psi_1^t) \Psi^t + (B_2 \circ \Psi_1^t)) + (N_3 \circ \Psi_1^t) \Psi^t \quad (22)$$

with

$$N_1 = \begin{pmatrix} 0 & A_1 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix}, \quad N_2 = \begin{pmatrix} 0 & 0 & 0 \\ 0 & 0 & A_2 \\ 0 & 0 & 0 \end{pmatrix}, \quad B_2 = \begin{pmatrix} 0 \\ b_2 \\ 0 \end{pmatrix},$$

$$N_3 = \begin{pmatrix} 0 & 0 & 0 \\ 0 & 0 & 0 \\ 0 & A_3 & 0 \end{pmatrix}.$$

The zero pattern of the matrices N_1, N_2, N_3 allows to compute explicit flow maps. Indeed, let $\psi_1^t, \psi_2^t, \psi_3^t$ be the flows of the differential equations

$$\begin{aligned} \frac{d}{dt}y(t) &= N_1 \cdot y(t), & \frac{d}{dt}y(t) &= N_2(y(t)) \cdot y(t) + B_2(y(t)), \\ \frac{d}{dt}y(t) &= N_3(y(t)) \cdot y(t). \end{aligned}$$

Then,

$$\begin{aligned} \psi_1^t(y) &= y + t \begin{pmatrix} A_1 \cdot y_2 \\ 0 \\ 0 \end{pmatrix}, & \psi_2^t(y) &= y + t \begin{pmatrix} 0 \\ A_2(y_1) \cdot y_3 + b_2(y_1) \\ 0 \end{pmatrix}, \\ \psi_3^t(y) &= y + t \begin{pmatrix} 0 \\ 0 \\ A_3(y_1) \cdot y_2 \end{pmatrix}. \end{aligned}$$

For $\tau > 0$, we obtain a first order splitting scheme for equation (22) by

$$\psi^\tau = \psi_2^\tau \circ \psi_1^\tau \circ \psi_3^\tau,$$

see, [28, §2.5]. Its adjoint is $\psi^{\tau*} = \psi_3^\tau \circ \psi_1^\tau \circ \psi_2^\tau$, and we can create a second order symmetric splitting method by

$$F_2^\tau = \psi^{\tau/2} \circ \psi^{\tau/2*} = \psi_2^{\tau/2} \circ \psi_1^{\tau/2} \circ \psi_3^\tau \circ \psi_1^{\tau/2} \circ \psi_2^{\tau/2}.$$

A corresponding fourth order splitting is obtained by

$$F_4^\tau = F_2^{\frac{\tau}{2-2^{1/3}}} \circ F_2^{\frac{-2^{1/3}\tau}{2-2^{1/3}}} \circ F_2^{\frac{\tau}{2-2^{1/3}}},$$

see [27, §4], and we obtain $\Psi^\tau - F_4^\tau = O(\tau^5)$ as $\tau \rightarrow 0$. By construction, the first $2d$ components of ψ^τ, F_2^τ , and F_4^τ , respectively, define symplectic maps on phase space.

3.4. The approximation scheme

At this point we have built up all the ingredients for computing an approximation to expectation values, which is fourth order accurate with respect to the semiclassical parameter ε . Let $a : \mathbb{R}^{2d} \rightarrow \mathbb{R}$ be a Schwartz function. According to the discussion in Section 3.3, we use time splitting schemes of order eight and order four, for the approximation of $a_0(t)$ and $a_2(t)$, respectively. That is,

$$a_0(t) = \tilde{a}_0^\tau(t) + O(\tau^8), \quad a_2(t) = \tilde{a}_2^\tau(t) + O(\tau^4),$$

as $\tau \rightarrow 0$, with $\tilde{a}_0^\tau(t) = a \circ (\phi^\tau \circ \dots \circ \phi^\tau)$ and

$$\begin{aligned} \tilde{a}_2(\tau) = & -\frac{1}{4} \left(\left(D^3 a \circ (\tilde{\Phi}^\tau \circ \dots \circ \tilde{\Phi}^\tau) \right)_{ijk} (\tilde{\Lambda}_{kji}^\tau \circ \dots \circ \tilde{\Lambda}_{kji}^\tau) \right. \\ & + 3 \left(D^2 a \circ (\tilde{\Phi}^\tau \circ \dots \circ \tilde{\Phi}^\tau) \right)_{ij} (\tilde{\Gamma}_{ji}^\tau \circ \dots \circ \tilde{\Gamma}_{ji}^\tau) \\ & \left. + \left(D a \circ (\tilde{\Phi}^\tau \circ \dots \circ \tilde{\Phi}^\tau) \right)_i (\tilde{\Xi}_i^\tau \circ \dots \circ \tilde{\Xi}_i^\tau) \right), \end{aligned}$$

where ϕ^τ is a symplectic eighth order splitting for the Hamiltonian flow Φ^τ , while $\tilde{\Phi}^\tau$ and the tensors $\tilde{\Lambda}^\tau, \tilde{\Gamma}^\tau, \tilde{\Xi}^\tau$ consist of the appropriate components of the fourth order splitting F_4^τ . Using the quasi-Monte Carlo estimate (21), we obtain

$$\begin{aligned} \langle \text{op}(a_0(t) + \varepsilon^2 a_2(t)) \psi_0, \psi_0 \rangle = & I^{N_0} (\tilde{a}_0^{\tau_0}(t)) + O((\log N_0)^{c_d} N_0^{-1}) + O(\tau_0^8) \\ & + \varepsilon^2 I^{N_2} (\tilde{a}_2^{\tau_2}(t)) + \varepsilon^2 O((\log N_2)^{c_d} N_2^{-1}) + \varepsilon^2 O(\tau_2^4) \end{aligned}$$

for $\tau_0, \tau_2 \rightarrow 0$ and $N_0, N_2 \rightarrow \infty$. Choosing the time steps and the number of sampling points such that

$$\max(\tau_0^8, (\log N_0)^{c_d} N_0^{-1}) \leq \varepsilon^4, \quad \max(\tau_2^4, (\log N_2)^{c_d} N_2^{-1}) \leq \varepsilon^2,$$

we finally obtain the desired asymptotic approximation

$$\langle \text{op}(a) \psi_t, \psi_t \rangle = I^{N_0} (\tilde{a}_0^{\tau_0}(t)) + \varepsilon^2 I^{N_2} (\tilde{a}_2^{\tau_2}(t)) + O(\varepsilon^4).$$

We note, that the number of sampling points N_2 can be chosen much smaller than N_0 and that the step size τ_2 can be chosen about the same size as τ_0 . Ignoring the logarithmic term in the quasi-Monte Carlo estimate, we deduce as a rule of thumb:

$$N_2 \approx \varepsilon^2 N_0, \quad \tau_2 \approx \tau_0.$$

Hence, although the system of differential equations for the approximation of the correction $a_2(t)$ is more intricate than the one for $a_0(t)$, for moderate dimensions d , the computation of the correction is less costly.

4. Numerical Experiments

For our numerical experiments, we consider the time dependent Schrödinger equation

$$i\varepsilon \partial_t \psi_t = \left(-\frac{\varepsilon^2}{2} \Delta + 2 - \cos(q_1) - \cos(q_2) \right) \psi_t$$

in two dimensions with torsional potential. The time interval is $[0, 15]$, and the initial data ψ_0 is chosen as a single Gaussian wave packet (20) with center $(q_0, p_0) = (1, 0.5, 0, 0)$. We validate the approach developed in Section 3 for different observables, namely the position and momentum operators given by

$$(q, p) \mapsto q_j, \quad (q, p) \mapsto p_j, \quad j = 1, 2$$

as well as the potential, kinetic and total energy operators defined by

$$(q, p) \mapsto V(q), \quad (q, p) \mapsto \frac{1}{2}|p|^2, \quad (q, p) \mapsto h(q, p)$$

Since the computation of the leading order Egorov term $a_0(t) = a \circ \Phi^t$ has already been elaborated in [10], we will mainly focus on the correction term $a_2(t)$. The main goal of our numerical experiments is to show that our suggested algorithm can reach order four accuracy with respect to ε , does this in an efficient way, and thus is feasible in a moderately high dimensional setting.

As a reference, we use grid-based solutions to the Schrödinger equation computed by a Strang splitting scheme with Fourier collocation, see [10, Appendix], and derive the needed expectation values. The parameters we use for computing the reference solution are given in Table 4.

Table 1. The discretization parameters for the grid-based reference solutions computed by a Strang splitting scheme with Fourier collocation.

ϵ	# time steps	domain	space grid
0.1	$1.2 * 10^5$	$[-3, 3] \times [-3, 3]$	1024×1024
0.05	$3.6 * 10^5$	$[-3, 3] \times [-3, 3]$	1024×1024
0.02	$3.6 * 10^5$	$[-3, 3] \times [-3, 3]$	1024×1024
0.01	$1.2 * 10^6$	$[-3, 3] \times [-3, 3]$	1024×1024

4.1. Time-evolution of observables

Figure 1 presents the expectation values computed by the full approximation

$$\langle \text{op}(a)\psi_t, \psi_t \rangle \approx I^{N_0}(\tilde{a}_0^{\tau_0}(t)) + \varepsilon^2 I^{N_2}(\tilde{a}_2^{\tau_2}(t)) \quad (23)$$

for the case $\varepsilon = 10^{-2}$. We note that the total energy is preserved, see also Remark 2.1. Figure 2 compares the expectation values computed from the reference solution with those from the Egorov approximation

$$\langle \text{op}(a)\psi_t, \psi_t \rangle \approx I^{N_0}(\tilde{a}_0^{\tau_0}(t)) \quad (24)$$

as well as the corrected approximation (23). We observe that the error of the corrected approach is of the order ε^4 , while the error of the uncorrected approximation (24) is only of the order ε^2 .

4.2. Asymptotic accuracy

To validate that the corrected algorithm is of the order of four in ε , we compute the maximal and mean deviation from our reference solution for $\varepsilon = 0.1, 0.05, 0.02, 0.01$, where the number of Halton points N_0 and N_2 and the size of the time steps τ_0 and τ_2 are given in Table 4.2. In Table 4.2 the computing times of the Egorov approximation scheme (24) and the correction term

$$\langle \text{op}(a_2(t))\psi_0, \psi_0 \rangle \approx I^{N_2}(\tilde{a}_2^{\tau_2}(t)) \quad (25)$$

are presented as well. For a given accuracy we can observe that, especially for small semiclassical parameters, (25) can be computed much faster than (24). In Figure 3 the expected fourth order of our approximation can be seen.

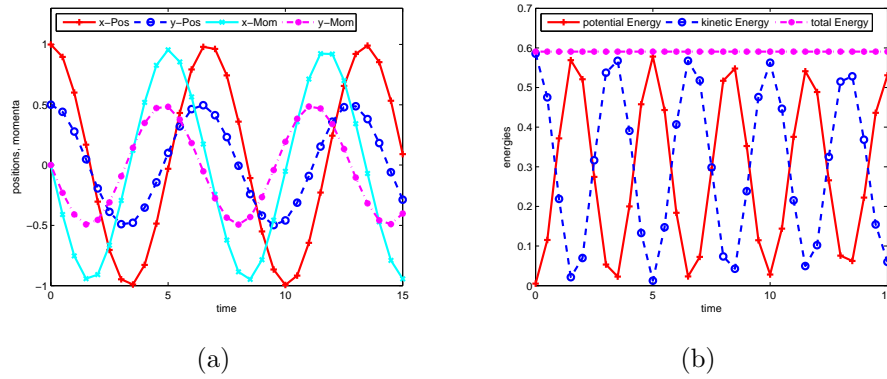


Figure 1. Figure 1(a) shows the expectation values of positions and momenta and Figure 1(b) the expectation values of kinetic, potential and total energy for the two dimensional torsion potential as a function of time computed by the approximation (23). The semiclassical parameter is chosen as $\varepsilon = 10^{-2}$.

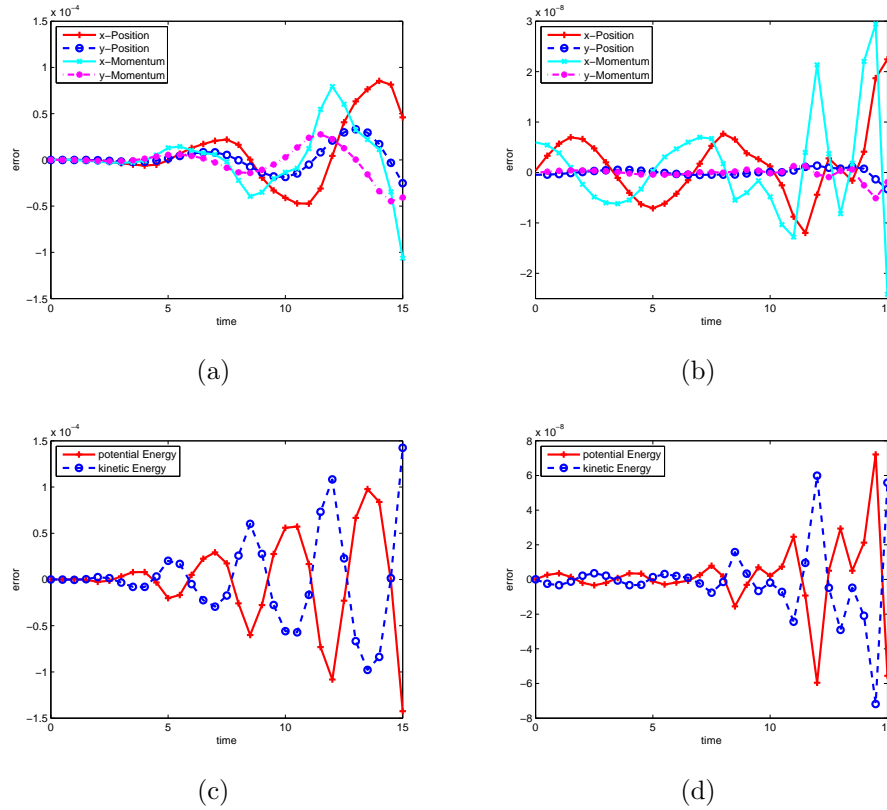


Figure 2. Figures 2(a) and 2(c) show the absolute error of the expectation values of positions and momenta as well of the kinetic and potential energy, respectively, using the simple Egorov approximation (24). Figures 2(b) and 2(d) show the same for the corrected approximation (23). The semiclassical parameter is chosen as $\varepsilon = 10^{-2}$. In the two pictures on the left, the error is of order ε^2 , whereas those on the right reach an order of ε^4 .

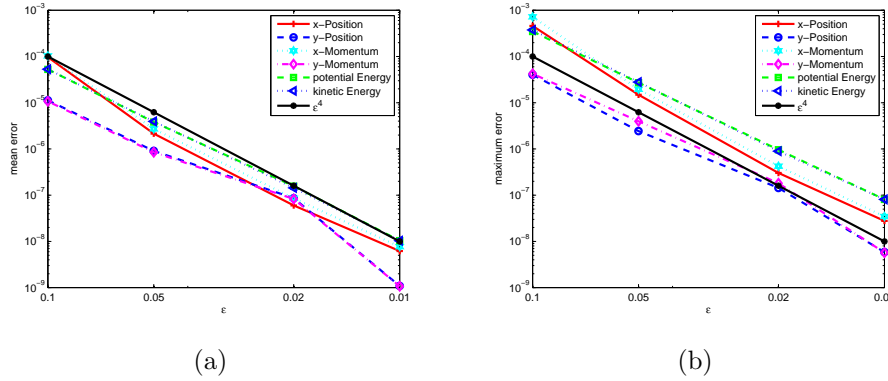


Figure 3. The mean (a) and maximal (b) error over time of the expectation values of positions and momenta, kinetic and potential energy computed with the corrected algorithm (23) as a function of ε . The values for the number of sampling points and the time step sizes are given in Table 4.2. Both plots show an order of ε^4 .

Table 2. Numbers of sampling points N_0 and N_2 , time step sizes τ_0 and τ_2 as well as the computing times t_0^{elaps} for the Egorov algorithm (24) and t_2^{elaps} for the corrections computed by the approximation scheme (25) that were used for the results in Figure 3.

ε	N_0	τ_0	t_0^{elaps}	N_2	τ_2	t_2^{elaps}
0.1	10^5	0.1	6.5 sec	500	2^{-2}	2.5 sec
0.05	10^6	0.1	46 sec	10^3	2^{-3}	6 sec
0.02	10^7	0.1	7.5 min	10^4	2^{-5}	2.5 min
0.01	10^8	0.1	65 min	10^4	2^{-5}	2.5 min

4.3. Discretization errors

For the experiments shown in Figure 4 and Figure 5 the values of the leading order approximation (24) are computed with $N_0 = 10^8$ sampling points and a time step size of $\tau_0 = 2^{-4}$. In Figure 4, we examine the dependency of the absolute error of the expectation values from the number of particles N_2 . To do so we compare expectation values coming from (23) with those from our reference and compute the maximal as well as the mean error over the time interval $[0, 15]$. For the computation of (25) we apply our fourth order splitting scheme with a time step size of $\tau_2 = 10^{-2}$. We can see that the mean as well as the maximal error decrease with an order slightly worse than ε^2/N_2 until they reach a lower bound of the order ε^4 . In Figure 5, the mean and maximal error over time of the approximated expectation values depending on the step size τ_2 is displayed. Here the number of sampling points is chosen as $N_2 = 10^4$. As expected, we observe that the mean as well as the maximum error decrease with an order of $\varepsilon^2\tau_2^4$ until they reach values of the order ε^4 .

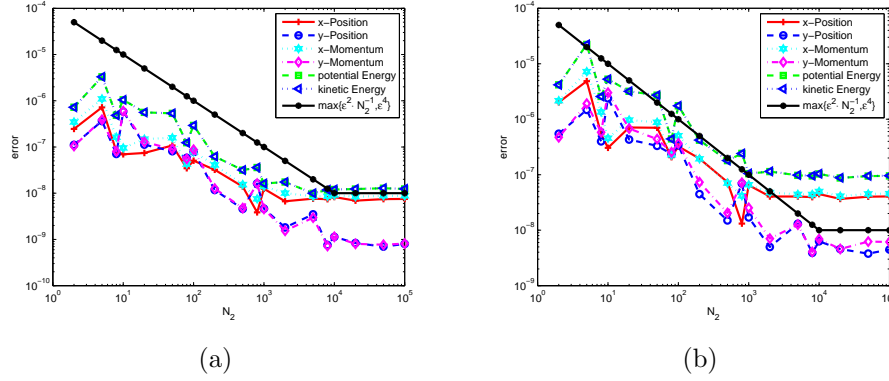


Figure 4. The mean (a) and maximal (b) error over time of the expectation values of positions and momenta, kinetic and potential energy computed with the approximation (23) as a function of N_2 . The semiclassical parameter is chosen as $\varepsilon = 10^{-2}$ and the time step for the splitting scheme $F_4^{T_2}$ as $\tau_2 = 2^{-4}$. In both plots the order of the error is slightly worse than ε^2/N_2 and bounded from below by values of the order ε^4 .

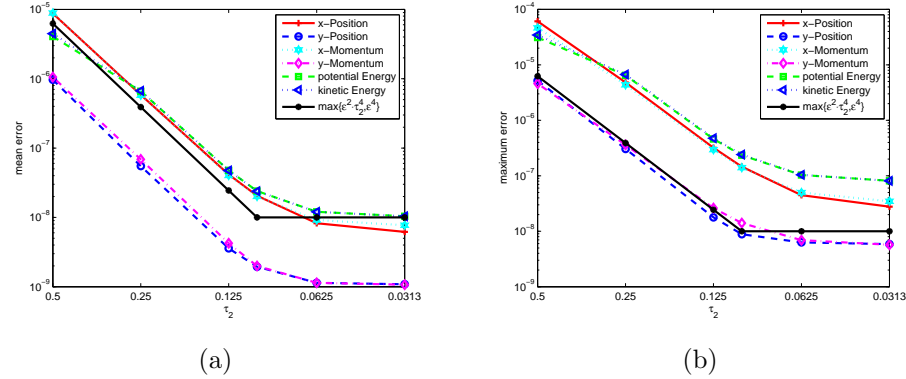


Figure 5. The mean (a) and maximal (b) error over time of the expectation values of positions and momenta, kinetic and potential energy computed with the corrected algorithm (23) as a function of τ_2 . The semiclassical parameter is chosen as $\varepsilon = 10^{-2}$ and the number of sampling points for (25) as $N_0 = 10^4$. In both plots the error is of the order $\varepsilon^2\tau_2^4$ but bounded from below by values of order ε^4 .

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Appendix A. Wigner functions and Weyl operators

Let $\varepsilon > 0$ be a small positive parameter. The ε -scaled Wigner function $W_\psi : \mathbb{R}^{2d} \rightarrow \mathbb{R}$ of a square integrable function $\psi \in L^2(\mathbb{R}^d)$ is defined as

$$W_\psi(q, p) = (2\pi\varepsilon)^{-d} \int_{\mathbb{R}^d} e^{ip \cdot y/\varepsilon} \psi(q - \tfrac{1}{2}y) \overline{\psi}(q + \tfrac{1}{2}y) dy, \quad (q, p) \in \mathbb{R}^{2d},$$

see for example [6, §1.8] or [7, §9]. Using Plancherel's theorem it can easily be shown that $\|\psi\| = 1$ implies

$$\int_{\mathbb{R}^{2d}} W_\psi(z) dz = 1.$$

The real-valued Wigner function W_ψ can therefore be interpreted as a phase space description of the wave function ψ . In contrast to classical phase space distributions, the Wigner function may also attain negative values. The Wigner function is in close relation to the ε -scaled Weyl quantization $\text{op}(a)$ of Schwartz functions $a : \mathbb{R}^{2d} \rightarrow \mathbb{R}$, since

$$\langle \psi, \text{op}(a)\psi \rangle = \int_{\mathbb{R}^{2d}} a(z) W_\psi(z) dz$$

with

$$(\text{op}(a)\psi)(q) = (2\pi\varepsilon)^{-d} \int_{\mathbb{R}^{2d}} a(\tfrac{1}{2}(q+y), p) e^{ip \cdot (q-y)/\varepsilon} \psi(y) dy$$

for $\psi \in L^2(\mathbb{R}^d)$. Weyl quantization also extends to unbounded phase space functions a , if operator domains are defined suitably. For example, $-\varepsilon^2 \Delta = \text{op}(|p|^2)$. The composition of Weyl operators is a Weyl operator $\text{op}(a)\text{op}(b) = \text{op}(c)$, and the symbol $c : \mathbb{R}^{2d} \rightarrow \mathbb{R}$ has an asymptotic expansion in powers of ε ,

$$c \sim \sum_{k \in \mathbb{N}} \left(\frac{\varepsilon}{2i} \right)^k \{a, b\}_k,$$

where the k th generalized Poisson bracket is defined as

$$\{a, b\}_k = \sum_{|\alpha|+|\beta|=k} \frac{(-1)^{|\beta|}}{\alpha! \beta!} \partial_q^\alpha \partial_p^\beta b \partial_q^\beta \partial_p^\alpha a, \quad (\text{A.1})$$

see [1, Appendix]. We note that the first generalized Poisson bracket coincides with the usual Poisson bracket, $\{a, b\}_1 = \partial_p a \partial_q b - \partial_q a \partial_p b$. Moreover, the antisymmetry of the Poisson bracket is also satisfied by its generalizations, if k is odd.

Lemma Appendix A.1. *Let $a, b : \mathbb{R}^{2d} \rightarrow \mathbb{R}$ be smooth functions and $k \in \mathbb{N}$. Then,*

$$\{a, b\}_k - \{b, a\}_k = \begin{cases} 0, & k \text{ even} \\ 2\{a, b\}_k, & k \text{ odd} \end{cases} \quad (\text{A.2})$$

Proof. By interchanging the multi-indices α and β in the formula for $\{g, f\}_k$ we get

$$\{a, b\}_k - \{b, a\}_k = \sum_{|\alpha|+|\beta|=k} \frac{(-1)^{|\beta|} - (-1)^{|\alpha|}}{\alpha! \beta!} \partial_q^\alpha \partial_p^\beta b \partial_q^\beta \partial_p^\alpha a.$$

Since $|\alpha| + |\beta| = k$, we conclude that for even k , $|\alpha|$ is even if and only if $|\beta|$ is even and for odd k , $|\alpha|$ is even if and only if $|\beta|$ is odd, which finishes the proof. \square

In consequence, the commutator of two Weyl operators has an asymptotic expansion in odd powers of ε ,

$$[\text{op}(a), \text{op}(b)] = \text{op}(a)\text{op}(b) - \text{op}(b)\text{op}(a) \sim 2 \sum_{k \in 2\mathbb{N}+1} \left(\frac{\varepsilon}{2i}\right)^k \text{op}(\{a, b\}_k).$$

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